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IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF

HIROYUKI KISO, ET AL.

SERIAL NO: 10/618,693

FILED: JULY 15, 2003

RCE FILED: NOVEMBER 23, 2005

FOR: CATALYST AND PROCESS FOR
PRODUCING A RIGID POLYURETHANE
FOAM

:

EXAMINER: COONEY, J. M.

:

GROUP ART UNIT: 1711

:

APPEAL BRIEF

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

SIR:

This is an appeal of the Final Rejection dated July 11, 2006 of Claims 1, 4, 29-33, 37 and 38. A Notice of Appeal was timely filed on October 10, 2006.

I. REAL PARTY IN INTEREST

The real party in interest in this appeal is Tosoh Corporation having an address 4560, Kaisei-cho, Shunan-shi, Yamaguchi-ken, 746-8501, Japan.

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II. RELATED APPEALS AND INTERFERENCES

Appellants, Appellants' legal representative and the assignee are aware of no appeals, interferences, or judicial proceedings which may be related to, directly affect or be directly affected by or have a bearing on the Board's decision in this appeal.

III. STATUS OF THE CLAIMS

Claims 1, 4, 29-33, 37 and 38 stand rejected and are herein appealed. Claims 5-28 stand withdrawn from consideration. Claims 2, 3, and 34-36 have been canceled.

IV. STATUS OF THE AMENDMENTS

An amendment under 37 CFR 41.33 is filed herewith. The amendment cancels Claims 34-36 and rewrites Claim 37 in independent form, without change in claim scope.

V. SUMMARY OF THE CLAIMED SUBJECT MATTER

Independent Claim 1 is drawn to a catalyst for producing a rigid polyurethane foam by means of at least one blowing agent selected from the group consisting of 1,1,1,3,3-pentafluoropropane (HFC-245fa), 1,1,1,3,3-pentafluorobutane (HFC-365mfc) and a low boiling point hydrocarbon, which comprises:

at least one amine compound (1) selected from the group consisting of dimethylethylamine, dimethylpropylamine, dimethylbutylamine, dimethylpentylamine, dimethylhexylamine, dimethylheptylamine, dimethyloctylamine, dimethylnonylamine, dimethyldecylamine, dimethylundecylamine, dimethyldodecylamine, dimethyltridecylamine, dimethyltetradecylamine, dimethylpentadecylamine and dimethylhexadecylamine,

and at least one amine compound (2) selected from the group consisting of triethylenediamine, N,N,N',N'-tetramethyl-1,6-hexanediamine and N,N-dimethylcyclohexylamine,

wherein said at least one amine compound (1) and said at least one amine compound (2) are present in amounts effective to improve at least one property of a rigid polyurethane foam produced by means of said at least one blowing agent and said catalyst, compared to an otherwise identical rigid polyurethane foam produced using said at least one amine compound (1) alone, or said at least one amine compound (2) alone, as the catalyst, and wherein said at least one property includes dimensional stability.

Independent Claim 37 is drawn to a catalyst for producing a rigid polyurethane foam by means of at least one blowing agent selected from the group consisting of 1,1,1,3,3-pentafluoropropane (HFC-245fa), 1,1,1,3,3-pentafluorobutane (HFC-365mfc) and a low boiling point hydrocarbon, which comprises:

at least one amine compound (1) selected from the group consisting of dimethylbutylamine, dimethylhexylamine, dimethyloctylamine, dimethylnonylamine, and dimethyldodecylamine,

and at least one amine compound (2) selected from the group consisting of triethylenediamine, N,N,N',N'-tetramethyl-1,6-hexanediamine and N,N-dimethylcyclohexylamine,

wherein said at least one amine compound (1) and said at least one amine compound (2) are present in amounts effective to improve at least one property of a rigid polyurethane foam produced by means of said at least one blowing agent and said catalyst, compared to an

otherwise identical rigid polyurethane foam produced using said at least one amine compound (1) alone, or said at least one amine compound (2) alone, as the catalyst, and wherein said at least one property includes dimensional stability.

Claim 37 is analogous to Claim 1, but it narrows the scope of the compound (1) Markush group.

Independent Claim 38 is drawn to a catalyst for producing a rigid polyurethane foam by means of at least one blowing agent selected from the group consisting of 1,1,1,3,3-pentafluoropropane (HFC-245fa), 1,1,1,3,3-pentafluorobutane (HFC-365mfc) and a low boiling point hydrocarbon, which comprises:

at least one amine compound (1) selected from the group consisting of dimethylethylamine, dimethylpropylamine, dimethylbutylamine, dimethylpentylamine, dimethylhexylamine, dimethylheptylamine, dimethyloctylamine, dimethylnonylamine, dimethyldecylamine, dimethylundecylamine, dimethyldodecylamine, dimethyltridecylamine, dimethyltetradecylamine, dimethylpentadecylamine and dimethylhexadecylamine,

and at least one amine compound (2) selected from the group consisting of triethylenediamine, N,N,N',N'-tetramethyl-1,6-hexanediamine and N,N-dimethylcyclohexylamine,

wherein said at least one amine compound (1) and said at least one amine compound (2) are present in amounts effective to improve a balance of properties of a rigid polyurethane foam produced by means of said at least one blowing agent and said catalyst, compared to an otherwise identical rigid polyurethane foam produced using said at least one amine compound (1) alone, or said at least one amine compound (2) alone, as the catalyst, and

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wherein said balance of properties is a combination of flowability, adhesive strength and dimensional stability.

Claim 38 is analogous to Claim 1, but the functional amounts are in reference to a balance of the recited properties.

Support for the above independent claims appear in original Claims 1 and 3, together with the specification at page 13, lines 2-13 and page 19, lines 10-22, with support for the recited amounts in Claims 1, 37 and 38 at page 12, lines 5-12, together with the comparative data in the specification beginning at page 53, line 17.

VI. GROUNDS OF REJECTION

Ground (A)

Claims 1, 4, 29-33 and 38 stand rejected under 35 U.S.C. § 103(a) as unpatentable over U.S. 4,585,804 (Lancaster et al) alone, or in view of U.S. 4,742,089 (Naka et al).

Ground (B)

Claims 1, 4, 29-38 stand rejected under 35 U.S.C. § 103(a) as unpatentable over U.S. 4,742,089 (Canaday et al) alone, or in view of Naka et al.

Ground (C)

Claims 1, 29-33 and 36-38 stand rejected under 35 U.S.C. § 112, first paragraph, as failing to satisfy the description requirement thereof.

VII. ARGUMENT

Preface to Grounds (A) and (B)

The present invention is based on Applicants' discovery that the combination of amine compounds (1) and (2) of the present claims produces a polyurethane foam that is superior to one using either amine compound alone as a catalyst when used in conjunction with a blowing agent selected from the group consisting of HFC-245fa, HFC-365mfc, and a low boiling point hydrocarbon.

Two sets of comparative data in the specification evidence this superiority, as discussed and set forth below.

In the first set of comparative data, which demonstrates the significance of using HFC-245fa or HFC-365mfc as the blowing agent, Examples 1-24 are according to the presently-claimed invention, and Comparative Examples 1-29 are not. The only difference between Comparative Examples 1-14 and the Examples is the catalyst used. Comparative Examples 15-26 employ the amine combination of the present invention, but with HCFC-141b as the blowing agent, i.e., a blowing agent other than HFC-245fa, HFC-365mfc, and a low boiling point hydrocarbon.

The data are shown in Tables 2, 4 and 6, at pages 58-59, 62, and 67, respectively. As described at page 68, lines 14-16, in each Example, a rigid urethane foam excellent in flowability, adhesive strength and dimensional stability, was obtained. For Comparative Examples 1-4 and 8-11, which used only amine (1) as the catalyst, large amounts of the catalyst were required, and the foams were inferior in flowability and dimensional stability, as described at page 68, lines 17-23. For Comparative Examples 5-7 and 12-14, which used only amine (2) as the catalyst, the foams were inferior in flowability, adhesive strength and

dimensional stability, as described at the paragraph bridging pages 68 and 69. For Comparative Examples 15-26, using the amine combination of the present claims but with a different blowing agent caused little change in flowability, adhesive strength or dimensional stability of the foams.

The second set of comparative data is analogous to the above-discussed first set, but highlights the significance of using a low boiling point hydrocarbon, such as cyclopentane alone or combined with isopentane, as an applicable blowing agent. See Tables 8, 10 and 12, and the descriptions of Examples 25-48, and Comparative Examples 30-58, in the specification at page 81, lines 1-26.

The comparative data is set forth in greater detail below. In the Tables, as can be confirmed by, for example, Table 1 at page 57 of the specification, Catalyst A is dimethylbutylamine; Catalyst B is dimethylhexylamine; Catalyst C is dimethyloctylamine; Catalyst D is dimethyldodecylamine; Catalyst E is triethylenediamine; Catalyst F is N,N,N',N'-tetramethyl-1,6-hexanediamine; and Catalyst G is N,N-dimethylcyclohexylamine.

When HFC-245fa was used as a blowing agent:

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm ²)	Dimensional stability (%)
Combination of Catalyst A and at least one of Catalysts E to G (Present Inv.)	Examples 1 to 3	81 to 82	1.17 to 1.20	-1.3 to -1.0
Catalyst A alone	Comparative Example 1	76	1.38	-8.8
Catalyst E alone	Comparative Example 5	72	0.45	-3.5
Catalyst F alone	Comparative Example 6	76	0.51	-5.2
Catalyst G alone	Comparative Example 7	76	0.48	-5.6

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm ²)	Dimensional stability (%)
Combination of Catalyst B and at least one of Catalysts E to G (Present Inv.)	Examples 4 to 6	79 to 81	1.32 to 1.45	-1.2 to -0.7
Catalyst B alone	Comparative Example 2	77	1.56	-9.2
Catalyst E alone	Comparative Example 5	72	0.45	-3.5
Catalyst F alone	Comparative Example 6	76	0.51	-5.2
Catalyst G alone	Comparative Example 7	76	0.48	-5.6

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm ²)	Dimensional stability (%)
Combination of Catalyst C and at least one of Catalysts E to G (Present Inv.)	Examples 7 to 9	79 to 83	1.21 to 1.33	-1.2 to -0.8
Catalyst C alone	Comparative Example 3	75	1.5	-8.5
Catalyst E alone	Comparative Example 5	72	0.45	-3.5
Catalyst F alone	Comparative Example 6	76	0.51	-5.2
Catalyst G alone	Comparative Example 7	76	0.48	-5.6

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm ²)	Dimensional stability (%)
Combination of Catalyst D and at least one of Catalysts E to G (Present Inv.)	Examples 10 to 12	79 to 83	1.14 to 1.28	-1.5 to -0.9
Catalyst D alone	Comparative Example 4	76	1.41	-7.7
Catalyst E alone	Comparative Example 5	72	0.45	-3.5
Catalyst F alone	Comparative Example 6	76	0.51	-5.2
Catalyst G alone	Comparative Example 7	76	0.48	-5.6

As is evident from the above tables, where HFC-245fa was used as a blowing agent, when Catalyst A, B, C or D was used alone, the dimensional stability was reduced substantially. Further, when Catalyst E, F or G was used alone, the adhesive strength and dimensional stability were reduced substantially. Accordingly, the adhesive strength and

dimensional stability can be improved by the combination of Catalyst A, B, C or D and Catalyst E, F or G in the present invention.

When HFC-365mfc was used as a blowing agent:

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm ²)	Dimensional stability (%)
Combination of Catalyst A and at least one of Catalysts E to G (Present Inv.)	Examples 13 to 15	78 to 80	1.26 to 1.48	-1.2 to -0.9
Catalyst A alone	Comparative Example 8	75	1.62	-8.2
Catalyst E alone	Comparative Example 12	72	0.67	-2.7
Catalyst F alone	Comparative Example 13	75	0.76	-4.3
Catalyst G alone	Comparative Example 14	76	0.72	-4.4

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm ²)	Dimensional stability (%)
Combination of Catalyst B and at least one of Catalysts E to G (Present Inv.)	Examples 16 to 18	78 to 79	1.42 to 1.66	-1.1 to -0.7
Catalyst B alone	Comparative Example 9	75	1.75	-7.5
Catalyst E alone	Comparative Example 12	72	0.67	-2.7
Catalyst F alone	Comparative Example 13	75	0.76	-4.3
Catalyst G alone	Comparative Example 14	76	0.72	-4.4

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm ²)	Dimensional stability (%)
Combination of Catalyst C and at least one of Catalysts E to G (Present Inv.)	Examples 19 to 21	78 to 80	1.30 to 1.52	-1.2 to -0.7
Catalyst C alone	Comparative Example 10	75	1.68	-7.8
Catalyst E alone	Comparative Example 12	72	0.67	-2.7
Catalyst F alone	Comparative Example 13	75	0.76	-4.3
Catalyst G alone	Comparative Example 14	76	0.72	-4.4

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm ²)	Dimensional stability (%)
Combination of Catalyst D and at least one of Catalysts E to G (Present Inv.)	Examples 22 to 24	78 to 81	1.25 to 1.43	-1.4 to -0.8
Catalyst D alone	Comparative Example 11	76	1.55	-7.2
Catalyst E alone	Comparative Example 12	72	0.67	-2.7
Catalyst F alone	Comparative Example 13	75	0.76	-4.3
Catalyst G alone	Comparative Example 14	76	0.72	-4.4

As is evident from the above tables, where HFC-365mfc was used as a blowing agent, when Catalyst A, B, C or D was used alone, the dimensional stability was reduced substantially. Further, when Catalyst E, F or G was used alone, the adhesive strength and dimensional stability were reduced substantially. Accordingly, the adhesive strength and

dimensional stability are improved by the combination of Catalyst A, B, C or D and Catalyst E, F or G in the present invention.

When a low boiling point hydrocarbon was used as a blowing agent:

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm ²)	Dimensional stability (%)
Combination of Catalyst A and at least one of Catalysts E to G (Present Inv.)	Examples 25 to 27, and 37 to 39	78 to 81	1.59 to 1.81	-1.2 to -0.8
Catalyst A alone	Comparative Examples 30 and 37	73 to 74	1.74 to 1.79	-7.7 to -6.8
Catalyst E alone	Comparative Examples 34 and 41	70	0.88 to 0.90	-3.1 to -2.8
Catalyst F alone	Comparative Examples 35 and 42	73 to 74	1.00 to 1.02	-4.6 to -4.3
Catalyst G alone	Comparative Examples 36 and 43	73 to 74	0.95 to 0.96	-5.0 to -4.3

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm ²)	Dimensional stability (%)
Combination of Catalyst B and at least one of Catalysts E to G (Present Inv.)	Examples 28 to 30, and 40 to 42	76 to 79	1.80 to 2.03	-1.1 to -0.5
Catalyst B alone	Comparative Examples 31 and 38	74 to 75	1.95 to 2.03	-8.1 to -7.2
Catalyst E alone	Comparative Examples 34 and 41	70	0.88 to 0.90	-3.1 to -2.8
Catalyst F alone	Comparative Examples 35 and 42	73 to 74	1.00 to 1.02	-4.6 to -4.3
Catalyst G alone	Comparative Examples 36 and 43	73 to 74	0.95 to 0.96	-5.0 to -4.3

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm ²)	Dimensional stability (%)
Combination of Catalyst C and at least one of Catalysts E to G (Present Inv.)	Examples 31 to 33, and 43 to 45	76 to 81	1.64 to 1.86	-1.1 to -0.6
Catalyst C alone	Comparative Examples 32 and 39	72 to 73	1.88 to 1.95	-7.5 to -6.8
Catalyst E alone	Comparative Examples 34 and 41	70	0.88 to 0.90	-3.1 to -2.8
Catalyst F alone	Comparative Examples 35 and 42	73 to 74	1.00 to 1.02	-4.6 to -4.3
Catalyst G alone	Comparative Examples 36 and 43	73 to 74	0.95 to 0.96	-5.0 to -4.3

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm ²)	Dimensional stability (%)
Combination of Catalyst D and at least one of Catalysts E to G (Present Inv.)	Examples 34 to 36, and 46 to 48	76 to 81	1.55 to 1.79	-1.4 to -0.7
Catalyst D alone	Comparative Examples 33 and 40	73 to 74	1.78 to 1.83	-6.8 to -6.0
Catalyst E alone	Comparative Examples 34 and 41	70	0.88 to 0.90	-3.1 to -2.8
Catalyst F alone	Comparative Examples 35 and 42	73 to 74	1.00 to 1.02	-4.6 to -4.3
Catalyst G alone	Comparative Examples 36 and 43	73 to 74	0.95 to 0.96	-5.0 to -4.3

As is evident from the above tables, where a low boiling point hydrocarbon was used as a blowing agent, when Catalyst A, B, C or D was used alone, the dimensional stability was reduced substantially. Further, when Catalyst E, F or G was used alone, the adhesive strength and dimensional stability were reduced substantially. Accordingly, the adhesive strength and dimensional stability can be improved by the combination of Catalyst A, B, C or D and Catalyst E, F or G in the present invention.

As described in the specification beginning at page 9, line 25, blowing agents of the type recited in the present claims are hardly soluble in a polyol as compared to dichloromonofluororethane (HCFC) analogs, are expensive, and if various tertiary amine catalysts are used, the amount of water to be used will be substantial, whereby there will be a problem that, compared to rigid polyurethane foams obtained using conventional HCFC or

the like as a blowing agent, the foam will be inferior in flowability, adhesive strength and dimensional stability. Thus, the presently-claimed invention is particularly efficacious when used to make rigid polyurethane foams using at least one of the blowing agents recited in the present claims.

Ground (A)

Claims 1, 4, 29-33 and 38 stand rejected under 35 U.S.C. § 103(a) as unpatentable over Lancaster et al alone, or in view of Naka et al. That rejection is untenable and should not be sustained.

Lancaster et al discloses “improved” rigid polyurethane foams produced in the presence of, *inter alia*, a blowing agent and at least one catalyst, wherein the improvement comprises incorporating a compound having at least one primary aliphatic amine group of a particular average molecular weight range (column 1, lines 35-56). Applicable catalysts listed include, *inter alia*, triethylene diamine and dimethylethylamine (paragraph bridging columns 3 and 4). While Lancaster et al discloses that mixtures of catalysts can be used, no advantage is disclosed from doing so.

Naka et al is drawn to the production of rigid polyurethane foams by reacting a polyol component having a special composition with an isocyanate component in the presence of a blowing agent, a reaction catalyst and a foam stabilizer having particular properties (Abstract). The Examiner relies on Naka et al’s listing of triethylene diamine, as well as dimethylcyclohexylamine and tetramethylhexamethylene diamine, finding them to be “equivalent.” Note, however, that among the blowing agents listed in Naka et al (column 4, lines 24-34), none are within the terms of the present claims.

The Examiner holds that it would have been obvious both to use the triethylene diamine and dimethylethylamine of Lancaster et al in combination, and to substitute the triethylene diamine with the “equivalent” dimethylcyclohexylamine or tetramethylhexamethylene diamine of Naka et al in that combination.

In reply, Naka et al does not disclose triethylene diamine, dimethylcyclohexylamine and tetramethylhexamethylene diamine, as necessarily “equivalent.” Rather, they are each listed simply as applicable in Naka et al’s reaction catalyst. More significantly, the applied prior art recognizes no benefit in using various catalysts in combination, compared to the use of individual catalysts alone.

Thus, the active claims are necessarily patentable, since they are inclusive only of catalysts which provide a rigid polyurethane foam having at least one improved property, as recited, such as dimensional stability, compared to the use of component (1) alone, or component (2) alone. It should not be necessary for Applicants to actually **show** superiority for all such compositions, since the prior art, as discussed above, recognizes no benefit from any combination of the above amines.

In sum, neither the presently-claimed invention, nor the above-discussed superior results, are disclosed or suggested by the applied prior art.

In the Final Rejection, the Examiner cites various precedent with regard to showings of unexpected results, and finds that Applicants have not demonstrated unexpected results in that they have not compared to the prior art relied on and that the showings are not commensurate in scope with the breadth of the claims.

In reply, what can be a more germane comparison when an invention is drawn to a combination of two components, where each component is known individually for a

particular utility, the prior art suggests no advantage in using them in combination, and the comparison is of the combination compared to each component alone? What prior art of record is closer? Indeed, the comparative data is more probative than the applied prior art since it relates to particular blowing agents recited in the claims. Compare *Ex parte Humber*, 217 USPQ 265 (Bd. Pat. App. & Inter. 1981) (comparative data showing the claimed chlorine-containing compounds to be unexpected over various (non-prior art) chlorine-containing isomers was accepted as more probative over prior art, drawn to non-chlorine containing analogs of the claimed compounds, asserted to be closest.) The applied prior art does not disclose these blowing agents.

With regard to whether the showing is commensurate in scope with the claims, the claims require that the relative amounts of the respective components (1) and (2) be in amounts effective to improve at least one property of a rigid polyurethane foam produced by means of a particular blowing agent and the particular combination of components (1) and (2), compared to identical subject matter but using component (1) alone or component (2) alone, and wherein the at least one property includes dimensional stability. The Examiner would presumably require submission of data for compositions of all possible relative amounts of components (1) and (2), suggesting that there are some combinations of component (1) and component (2) that do not result in an improved property, such as amounts wherein one of the components is present in an infinitesimal amount.

If that is the Examiner's position, Applicants submit that since they have already shown that there are such compositions that do result in an improvement in property, it would be a matter of routine experimentation to determine numerical percentage ranges in which such improved properties are obtained. It would defeat the Constitutional goal of advancing

progress in the useful arts to delay early disclosure of a meritorious invention by conducting nothing more than routine experimentation.

Claim 29

Claim 29 is separately patentable, because the combination of Lancaster et al and Naka et al could not have predicted the above-discussed superior results when using triethylenediamine as amine compound (2).

Claim 30

Claim 30 is separately patentable, because the combination of Lancaster et al and Naka et al could not have predicted the above-discussed superior results when using N,N,N',N'-tetramethyl-1,6-hexanediamine as amine compound (2).

Claim 31

Claim 31 is separately patentable, because the combination of Lancaster et al and Naka et al could not have predicted the above-discussed superior results when using N,N-dimethylcyclohexylamine as amine compound (2).

Claim 38

Claim 38 is separately patentable, since the applied prior art does not suggest catalysts which provide a rigid polyurethane foam having an improved balance of flowability, adhesive strength and dimensional stability, compared to the use of component (1) alone, or component (2) alone.

For all the above reasons, it is respectfully requested that the rejection be
REVERSED.

Ground (B)

Claims 1, 4, and 29-38 stand rejected under 35 U.S.C. § 103(a) as unpatentable over Canaday et al alone, or in view of Naka et al. That rejection is untenable and should not be sustained.

Canaday et al discloses polyurethane foams derived from, *inter alia*, at least one halogenated organic blowing agent and at least one amine catalyst (column 2, lines 13-25). While Canaday et al discloses that any of the blowing agents generally employed in similar prior art foam products may be employed, Canaday et al lists only chlorine-containing blowing agents (paragraph bridging columns 10 and 11). As applicable amines, Canaday et al lists tertiary amines such as triethylenediamine and dimethylbutylamine (paragraph bridging columns 11 and 12). While Canaday et al discloses that mixtures of the amines may be used (column 12, lines 3-4), no advantage is disclosed from doing so.

Naka et al has been discussed above under Ground (A).

The Examiner, in effect, employs Canaday et al in the same way as Lancaster et al is applied, as discussed above under Ground (A). Applicants' response in Ground (A) is the same herein, except that Canaday et al replaces Lancaster et al.

Claim 29

Claim 29 is separately patentable, because the combination of Canaday et al and Naka et al could not have predicted the above-discussed superior results when using triethylenediamine as amine compound (2).

Claim 30

Claim 30 is separately patentable, because the combination of Canaday et al and Naka et al could not have predicted the above-discussed superior results when using N,N,N',N'-tetramethyl-1,6-hexanediamine as amine compound (2).

Claim 31

Claim 31 is separately patentable, because the combination of Canaday et al and Naka et al could not have predicted the above-discussed superior results when using N,N-dimethylcyclohexylamine as amine compound (2).

Claim 37

Claim 37 is separately patentable, because the combination of Canaday et al and Naka et al could not have predicted the above-discussed superior results when using any of the amines in the component (1) Markush group, together with any of the amines in the component (2) Markush group.

Claim 38

Claim 38 is separately patentable, since the applied prior art does not suggest catalysts which provide a rigid polyurethane foam having an improved balance of flowability, adhesive strength and dimensional stability, compared to the use of component (1) alone, or component (2) alone.

For all the above reasons, it is respectfully requested that the rejection be
REVERSED.

Ground (C)

Claims 1, 29-33 and 36-38 stand rejected under 35 U.S.C. § 112, first paragraph, as failing to satisfy the description requirement therein. That rejection is untenable and should not be sustained.

The Examiner finds that the recital of the at least one amine compound (1) and the at least one amine compound (2) in functional effective amounts, as recited at the end of Claims 1 and 38, is not described in the specification.

In reply, the issue is whether Applicants had possession of the invention now claimed as of the filing date. Indeed, the epitome of the embodiment of the active claims is Applicants' discovery that when using any of the blowing agents recited in the claims, the recited catalyst, i.e., the combination of components (1) and (2), produces superior results not obtained by using component (1) alone, or component (2) alone. This discovery is borne out by the above-discussed comparative data of record, wherein Applicants actually describe the differences in results. See, for example, the description at page 68, lines 17-23, wherein

deficiencies resulting from the use of amine compound (1) are described, and the following paragraph, i.e., bridging pages 68 and 69, wherein deficiencies resulting from the use of amine compound (2) alone are described, compared to the combination of components (1) and (2).

For all the above reasons, it is respectfully requested that the rejection be
REVERSED.

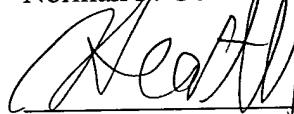
VIII. CONCLUSION

For the above reasons, it is respectfully requested that all the rejections still pending be REVERSED.

Respectfully submitted,

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HAP/rle

CLAIMS APPENDIX

Claim 1: A catalyst for producing a rigid polyurethane foam by means of at least one blowing agent selected from the group consisting of 1,1,1,3,3-pentafluoropropane (HFC-245fa), 1,1,1,3,3-pentafluorobutane (HFC-365mfc) and a low boiling point hydrocarbon, which comprises:

at least one amine compound (1) selected from the group consisting of dimethylethylamine, dimethylpropylamine, dimethylbutylamine, dimethylpentylamine, dimethylhexylamine, dimethylheptylamine, dimethyloctylamine, dimethylnonylamine, dimethyldecylamine, dimethylundecylamine, dimethyldodecylamine, dimethyltridecylamine, dimethyltetradecylamine, dimethylpentadecylamine and dimethylhexadecylamine,

and at least one amine compound (2) selected from the group consisting of triethylenediamine, N,N,N',N'-tetramethyl-1,6-hexanediamine and N,N-dimethylcyclohexylamine,

wherein said at least one amine compound (1) and said at least one amine compound (2) are present in amounts effective to improve at least one property of a rigid polyurethane foam produced by means of said at least one blowing agent and said catalyst, compared to an otherwise identical rigid polyurethane foam produced using said at least one amine compound (1) alone, or said at least one amine compound (2) alone, as the catalyst, and wherein said at least one property includes dimensional stability.

Claim 4: The catalyst for producing a rigid polyurethane foam according to Claim 1, wherein the composition of the at least one amine compound (1) and the at least one amine compound (2) comprises from 10 to 95 wt% of the at least one amine compound (1) and from 90 to 5 wt% of the at least one amine compound (2).

Claim 29: The catalyst for producing a rigid polyurethane foam according to Claim 1, wherein the at least one amine compound (2) comprises triethylenediamine.

Claim 30: The catalyst for producing a rigid polyurethane foam according to Claim 1, wherein the at least one amine compound (2) comprises N,N,N',N'-tetramethyl-1,6-hexanediamine.

Claim 31: The catalyst for producing a rigid polyurethane foam according to Claim 1, wherein the at least one amine compound (2) comprises N,N-dimethylcyclohexylamine.

Claim 32: The catalyst for producing a rigid polyurethane foam according to Claim 1, which additionally comprises at least one other catalyst.

Claim 33: The catalyst for producing a rigid polyurethane foam according to Claim 32, wherein the at least one other catalyst is selected from the group consisting of organic metal catalysts, metal carboxylate catalysts, tertiary amine catalysts other than amine compounds (1) and other than amine compounds (2), and quaternary ammonium salt catalysts.

Claim 34: A catalyst for producing a rigid polyurethane foam by means of at least one blowing agent selected from the group consisting of 1,1,1,3,3-pentafluoropropane (HFC-245fa), 1,1,1,3,3-pentafluorobutane (HFC-365mfc) and a low boiling point hydrocarbon, which comprises:

at least one amine compound (1) selected from the group consisting of dimethylpropylamine, dimethylbutylamine, dimethylpentylamine, dimethylhexylamine,

dimethylheptylamine, dimethyloctylamine, dimethylnonylamine, dimethyldecylamine, dimethylundecylamine, dimethyldodecylamine, dimethyltridecylamine, dimethyltetradecylamine, dimethylpentadecylamine and dimethylhexadecylamine, and at least one amine compound (2) selected from the group consisting of triethylenediamine, N,N,N',N'-tetramethyl-1,6-hexanediamine and N,N-dimethylcyclohexylamine.

Claim 35: The catalyst for producing a rigid polyurethane foam according to Claim 34, wherein the at least one amine compound (1) is selected from the group consisting of dimethylbutylamine, dimethylhexylamine, dimethyloctylamine, and dimethyldodecylamine.

Claim 36: The catalyst for producing a rigid polyurethane foam according to Claim 34, wherein said at least one amine compound (1) and said at least one amine compound (2) are present in amounts effective to improve at least one property of a rigid polyurethane foam produced by means of said at least one blowing agent and said catalyst, compared to an otherwise identical rigid polyurethane foam produced using said at least one amine compound (1) alone, or said at least one amine compound (2) alone, as the catalyst, and wherein said at least one property includes dimensional stability.

Claim 37: The catalyst for producing a rigid polyurethane foam according to Claim 35, wherein said at least one amine compound (1) and said at least one amine compound (2) are present in amounts effective to improve at least one property of a rigid polyurethane foam produced by means of said at least one blowing agent and said catalyst, compared to an otherwise identical rigid polyurethane foam produced using said at least one amine

compound (1) alone, or said at least one amine compound (2) alone, as the catalyst, and wherein said at least one property includes dimensional stability.

Claim 38: A catalyst for producing a rigid polyurethane foam by means of at least one blowing agent selected from the group consisting of 1,1,1,3,3-pentafluoropropane (HFC-245fa), 1,1,1,3,3-pentafluorobutane (HFC-365mfc) and a low boiling point hydrocarbon, which comprises:

at least one amine compound (1) selected from the group consisting of dimethylethylamine, dimethylpropylamine, dimethylbutylamine, dimethylpentylamine, dimethylhexylamine, dimethylheptylamine, dimethyloctylamine, dimethylnonylamine, dimethyldecylamine, dimethylundecylamine, dimethyldodecylamine, dimethyltridecylamine, dimethyltetradecylamine, dimethylpentadecylamine and dimethylhexadecylamine,

and at least one amine compound (2) selected from the group consisting of triethylenediamine, N,N,N',N'-tetramethyl-1,6-hexanediamine and N,N-dimethylcyclohexylamine,

wherein said at least one amine compound (1) and said at least one amine compound (2) are present in amounts effective to improve a balance of properties of a rigid polyurethane foam produced by means of said at least one blowing agent and said catalyst, compared to an otherwise identical rigid polyurethane foam produced using said at least one amine compound (1) alone, or said at least one amine compound (2) alone, as the catalyst, and wherein said balance of properties is a combination of flowability, adhesive strength and dimensional stability.

EVIDENCE APPENDIX

The comparative data described in the specification beginning at page 53, line 17
through page 82, line 5.

a low boiling point hydrocarbon, is used as a blowing agent, it is possible to produce a rigid polyisocyanurate foam excellent in the flame retardancy, adhesive strength and dimensional stability of the foam, without impairing 5 the physical properties of the foam. Further, according to the process of the present invention, it is possible to obtain a rigid polyisocyanurate foam having physical properties comparable to a foam produced by a conventional process.

10 Now, with reference to Examples and Comparative Examples, the catalyst for producing a rigid polyurethane foam and the process for producing a rigid polyurethane foam by means of such a catalyst, will be described. However, it should be understood that the present 15 invention is no by means restricted by such specific Examples.

In the following Examples and Comparative Examples, the various properties were measured by the following measuring methods.

20 •Measured items for the reactivity
Cream time: The time until the foam starts to rise was measured by visual observation.

Gel time: As the reaction proceeds, the time until the liquid substance changes to a resinous substance, was 25 measured.

Tack free time: The time until tackiness of the foam surface disappears, was measured.

Rise time: The time until the rise of the foam stops, was measured by visual observation.

•Flowability of foam: A predetermined amount of a mixed liquid was injected into an aluminum mold of 5 100×25×3.0 cm, and the length (cm) of the formed foam was measured. The longer the length of the foam, the better the flowability.

•Core density of foam:

Free foaming was carried out by means of an aluminum 10 mold of 50×50×4.5 cm, and the center portion of the formed foam was cut into a size of 20×20×3 cm, whereupon the size and weight were accurately measured, and core density was calculated.

•Adhesive strength of foam:

15 Forming was carried out by setting a zinc-lined iron plate of 5×5 cm on the upper side of an aluminum mold of 25×25×8.0 cm. After foaming for 1 hour, the peel strength at 90° of the set iron plate was measured and taken as the adhesive strength of the foam.

20 •Dimensional stability of foam:

A foam foamed in an aluminum mold of 50×50×4.5 cm was maintained under a condition of -30°C × 48 hours, whereby a change in the thickness direction was measured.

EXAMPLES 1 to 12

25 A polyol, a blowing agent and a foam stabilizer were mixed in a blend ratio as identified in Table 1 to prepare Premix A. 47.1 g of Premix A was taken into a

300 ml polyethylene cup, and a catalyst as identified in Table 1 was added in an amount such that the reactivity as represented by the following gel time would be 90 seconds, whereupon the temperature was adjusted to 10°C.

5 A polyisocyanate liquid (MR-200) having the temperature adjusted to 10°C in a separate container, was put into the cup of Premix A in such an amount that the isocyanate index [isocyanate groups/OH groups (molar ratio) × 100)] would be 110, and immediately stirred by a stirrer at

10 6,500 rpm for 5 seconds. The mixed and stirred liquid was transferred to a 2 liter polyethylene cup having the temperature adjusted to 40°C, and the reactivity during foaming was measured. Then, the starting material scale was increased, and in the same manner, the mixed liquid

15 was put into a mold having the temperature adjusted to 40°C, whereupon foam molding was carried out. Upon expiration of 10 minutes from the introduction of the mixed liquid, the foam was removed from the mold. From the molded foam, the flowability, core density, adhesive

20 strength and dimensional stability of the foam were evaluated. The results are shown in Table 2.

Table 1

Table 1 (continued)

	Blend (parts by weight)	Examples								
		16	17	18	19	20	21	22	23	24
Polyol ¹⁾	100	100	100	100	100	100	100	100	100	100
HFC-245fa										
Premix A										
HFC-365mfc	30	30	30	30	30	30	30	30	30	30
Water	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5
Foam stabilizer ²⁾	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0
Catalyst A ³⁾										
Catalyst B ⁴⁾	0.6	0.4	0.5							
Catalyst C ⁵⁾										
Catalyst D ⁶⁾										
Catalyst E ⁷⁾	1.2									
Catalyst F ⁸⁾		0.8								
Catalyst G ⁹⁾			1.0							
Isocyanate INDEX ¹⁰⁾	110	110	110	110	110	110	110	110	110	110

1) Sucrose/aromatic amine type polyether polyol (OH value=440 mgKOH/g, manufactured by Asahi Glass Company, Limited)

2) Silicone type surfactant (tradename: SZ-1627), manufactured by Nippon Unicar Co., Ltd.

3) N,N-dimethylbutylamine (manufactured by Aldrich Co.)

4) N,N-dimethylhexylamine (manufactured by Aldrich Co.)

5) N,N-dimethyloctylamine (manufactured by Tokyo Kasei K.K.)

6) N,N-dimethyldecylamine (manufactured by Tokyo Kasei K.K.)

7) Dipropylene glycol solution containing 33 wt% of triethylenediamine (TEDA-L33, manufactured by TOSOH CORPORATION)

8) N,N,N',N'-tetramethyl-1,6-hexanediamine (TOYOCAT-MR, manufactured by TOSOH CORPORATION)

9) N,N-dimethylcyclohexylamine (manufactured by Aldrich Co.)

10) Crude MDI (MR-200), manufactured by Nippon Polyurethane K.K., INDEX=(mols of NCO groups/mols of OH groups) $\times 100$

Table 2

	Examples														
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
Reactivity (sec)															
Cream time	25	20	21	24	20	21	26	20	20	25	21	21	30	24	26
Gel time	90	91	90	89	90	90	91	90	89	90	90	91	90	90	90
Tack free time	129	120	125	128	122	126	128	122	125	126	123	125	130	122	124
Rise time	144	135	140	142	137	141	143	135	139	139	136	138	148	137	143
Physical properties of foam															
Flowability (cm)	81	82	79	81	80	79	83	82	79	83	82	78	80	79	79
Core density (kg/m ³)	28.3	27.3	27.4	28.3	27.4	27.5	28.1	27.2	27.4	28.4	27.5	27.3	29.5	28.5	28.6
Adhesive strength (kgf/cm ²)	1.17	1.29	1.20	1.32	1.45	1.34	1.21	1.33	1.23	1.14	1.28	1.22	1.26	1.48	1.40
Dimensional stability (%)	-1.0	-1.3	-1.2	-0.7	-1.2	-1.1	-0.8	-1.1	-1.2	-0.9	-1.3	-1.5	-0.9	-1.2	-1.2

Table 2 (continued)

	Examples								
	16	17	18	19	20	21	22	23	24
Reactivity (sec)									
Cream time	29	25	26	32	24	25	31	26	26
Gel time	89	91	91	91	89	90	90	90	90
Tack free time	128	123	127	130	124	126	128	125	128
Rise time	145	138	141	149	137	140	146	138	143
Physical properties of foam									
Flowability (cm)	78	79	79	78	80	79	78	81	80
Core density (kg/m ³)	29.5	28.6	28.3	29.3	28.4	28.7	29.6	28.7	28.6
Adhesive strength (kgf/cm ²)	1.42	1.66	1.57	1.30	1.52	1.44	1.25	1.43	1.34
Dimensional stability (%)	-0.7	-1.1	-1.0	-0.7	-1.0	-1.2	-0.8	-1.2	-1.4

COMPARATIVE EXAMPLES 1 to 7

A polyol, a blowing agent and a foam stabilizer were mixed in a blend ratio as identified in Table 3 to prepare Premix A. 47.1 g of Premix A was taken into a 5 300 ml polyethylene cup, and a catalyst as identified in Table 3 was added in an amount such that the reactivity as represented by the following gel time would be 90 seconds, whereupon the temperature was adjusted to 10°C. A polyisocyanate liquid (MR-200) having the temperature 10 adjusted to 10°C in a separate container, was put into the cup of Premix A in such an amount that the isocyanate index [isocyanate groups/OH groups (molar ratio) $\times 100$] would be 110, and immediately stirred by a stirrer at 6,500 rpm for 5 seconds. The mixed and stirred liquid 15 was transferred to a 2 liter polyethylene cup having the temperature adjusted to 40°C, and the reactivity during foaming was measured. Then, the starting material scale was increased, and in the same manner, the mixed liquid was put into a mold having the temperature adjusted to 20 40°C, whereupon foam molding was carried out. Upon expiration of 10 minutes from the introduction of the mixed liquid, the foam was removed from the mold. From the molded foam, the flowability, core density, adhesive strength and dimensional stability of the foam were 25 evaluated. The results are shown in Table 4.

Table 3

	Blend (parts by weight)	Comparative Examples													
		1	2	3	4	5	6	7	8	9	10	11	12	13	14
Polyol ¹⁾	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
HFC-245fa	30	30	30	30	30	30	30	30	30	30	30	30	30	30	30
HFC-365mfc															
Water	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5
Foam stabilizer ²⁾	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0
Catalyst A ³⁾	3.0														
Catalyst B ⁴⁾		3.5													
Catalyst C ⁵⁾			4.0												
Catalyst D ⁶⁾				4.8											
Catalyst E ⁷⁾					2.9										
Catalyst F ⁸⁾						2.1									
Catalyst G ⁹⁾							2.8								
Isocyanate INDEX ¹⁰⁾	110	110	110	110	110	110	110	110	110	110	110	110	110	110	110

1) Sucrose/aromatic amine type polyether polyol (OH value=440 mgKOH/g, manufactured by Asahi Glass Company, Limited)

2) Silicone type surfactant (tradename: SZ-1627), manufactured by Nippon Unicar Co., Ltd.

3) N,N-dimethylbutylamine (manufactured by Aldrich Co.)

4) N,N-dimethylhexylamine (manufactured by Aldrich Co.)

5) N,N-dimethyloctylamine (manufactured by Tokyo Kasei K.K.)

6) N,N-dimethyldecylamine (manufactured by Tokyo Kasei K.K.)

7) Dipropylene glycol solution containing 33 wt% of triethylenediamine (TEDA-L33, manufactured by TOSOH CORPORATION)

8) N,N,N',N'-tetramethyl-1,6-hexanediamine (TOYOCAT-MR, manufactured by Aldrich Co.)

9) N,N-dimethylcyclohexylamine (manufactured by Nippon Polyurethane K.K., INDEX=(mols of NCO groups/mols of OH groups) × 100

10) Crude MDI (MR-200), manufactured by Nippon Polyurethane K.K., INDEX=(mols of NCO

Table 4

	Comparative Examples													
	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Reactivity (sec)														
Cream time	20	20	21	21	25	20	20	24	23	25	26	30	24	25
Gel time	90	90	90	90	90	90	91	90	89	90	90	90	90	91
Tack free time	160	138	143	137	129	130	127	168	145	150	144	136	137	133
Rise time	141	147	154	153	143	142	140	155	154	162	160	150	148	146
Physical properties of foam														
Flowability (cm)	76	77	75	76	72	76	76	75	75	76	72	75	76	76
Core density (kg/m ³)	27.5	27.4	27.5	27.3	28.6	27.4	27.5	28.6	28.5	28.4	28.6	29.7	28.5	28.6
Adhesive strength (kgf/cm ²)	1.38	1.56	1.50	1.41	0.45	0.51	0.48	1.62	1.75	1.68	1.55	0.67	0.76	0.72
Dimensional stability (%)	-8.8	-9.2	-8.5	-7.7	-3.5	-5.2	-5.6	-8.2	-7.5	-7.8	-7.2	-2.7	-4.3	-4.4

EXAMPLES 13 to 24

A polyol, a blowing agent and a foam stabilizer were mixed in a blend ratio as identified in Table 1 to prepare Premix A. 47.1 g of Premix A was taken into a 5 300 ml polyethylene cup, and a catalyst as identified in Table 1 was added in an amount such that the reactivity as represented by the following gel time would be 90 seconds, whereupon the temperature was adjusted to 10°C. A polyisocyanate liquid (MR-200) having the temperature 10 adjusted to 10°C in a separate container, was put into the cup of Premix A in such an amount that the isocyanate index [isocyanate groups/OH groups (molar ratio) × 100] would be 110, and immediately stirred by a stirrer at 6,500 rpm for 5 seconds. The mixed and stirred liquid 15 was transferred to a 2 liter polyethylene cup having the temperature adjusted to 40°C, and the reactivity during foaming was measured. Then, the starting material scale was increased, and in the same manner, the mixed liquid was put into a mold having the temperature adjusted to 20 40°C, whereupon foam molding was carried out. Upon expiration of 10 minutes from the introduction of the mixed liquid, the foam was removed from the mold. From the molded foam, the flowability, core density, adhesive strength and dimensional stability of the foam were 25 evaluated. The results are shown in Table 2.

COMPARATIVE EXAMPLES 8 to 14

A polyol, a blowing agent and a foam stabilizer were

mixed in a blend ratio as identified in Table 3 to prepare Premix A. 47.1 g of Premix A was taken into a 300 ml polyethylene cup, and a catalyst as identified in Table 3 was added in an amount such that the reactivity 5 as represented by the following gel time would be 90 seconds, whereupon the temperature was adjusted to 20°C. A polyisocyanate liquid (MR-200) having the temperature adjusted to 20°C in a separate container, was put into the cup of Premix A in such an amount that the isocyanate 10 index [isocyanate groups/OH groups (molar ratio) × 100] would be 110, and immediately stirred by a stirrer at 6,500 rpm for 5 seconds. The mixed and stirred liquid was transferred to a 2 liter polyethylene cup having the temperature adjusted to 40°C, and the reactivity during 15 foaming was measured. Then, the starting material scale was increased, and in the same manner, the mixed liquid was put into a mold having the temperature adjusted to 40°C, whereupon foam molding was carried out. Upon expiration of 10 minutes from the introduction of the 20 mixed liquid, the foam was removed from the mold. From the molded foam, the flowability, core density, adhesive strength and dimensional stability of the foam were evaluated. The results are shown in Table 4.

COMPARATIVE EXAMPLES 15 to 29

25 A polyol, a blowing agent and a foam stabilizer were mixed in a blend ratio as identified in Table 5 to prepare Premix A. 47.1 g of Premix A was taken into a

300 ml polyethylene cup, and a catalyst as identified in Table 5 was added in an amount such that the reactivity as represented by the following gel time would be 90 seconds, whereupon the temperature was adjusted to 20°C.

5 A polyisocyanate liquid (MR-200) having the temperature adjusted to 20°C in a separate container, was put into the cup of Premix A in such an amount that the isocyanate index [isocyanate groups/OH groups (molar ratio) $\times 100$] would be 110, and immediately stirred by a stirrer at

10 6,500 rpm for 5 seconds. The mixed and stirred liquid was transferred to a 2 liter polyethylene cup having the temperature adjusted to 40°C, and the reactivity during foaming was measured. Then, the starting material scale was increased, and in the same manner, the mixed liquid

15 was put into a mold having the temperature adjusted to 40°C, whereupon foam molding was carried out. Upon expiration of 10 minutes from the introduction of the mixed liquid, the foam was removed from the mold. From the molded foam, the flowability, core density, adhesive

20 strength and dimensional stability of the foam were evaluated. The results are shown in Table 6.

Table 5

	Blend (parts by weight)	Comparative Examples														
		15	16	17	18	19	20	21	22	23	24	25	26	27	28	29
Polyol ¹⁾	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
HCFC-141b	25	25	25	25	25	25	25	25	25	25	25	25	25	25	25	25
Water	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5
Foam stabilizer ²⁾	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0
Catalyst A ³⁾	0.4	0.3	0.4													
Catalyst B ⁴⁾				0.5	0.35	0.45										
Catalyst C ⁵⁾							0.55	0.35	0.5							
Catalyst D ⁶⁾								1.1				0.65	0.45	0.6		
Catalyst E ⁷⁾	0.8				1.0				0.7			1.3				
Amine Catalyst F ⁸⁾		0.6				0.7				0.9			0.9			
Amine Catalyst G ⁹⁾			0.8				0.9				1.0			1.2		
Isocyanate INDEX ¹⁰⁾	110	110	110	110	110	110	110	110	110	110	110	110	110	110	110	110

- 1) Sucrose/aromatic amine type polyether polyol (OH value=440 mgKOH/g, manufactured by Asahi Glass Company, Limited)
- 2) Silicone type surfactant (tradename: SZ-1627), manufactured by Nippon Unicar Co., Ltd.
- 3) N,N-dimethylbutylamine (manufactured by Aldrich Co.)
- 4) N,N-dimethylhexylamine (manufactured by Aldrich Co.)
- 5) N,N-dimethyloctylamine (manufactured by Tokyo Kasei K.K.)
- 6) N,N-dimethyldodecylamine (manufactured by Tokyo Kasei K.K.)
- 7) Dipropylene glycol solution containing 33 wt% of triethylenediamine (TEDA-L33, manufactured by TOSOH CORPORATION)
- 8) N,N,N',N'-tetramethyl-1,6-hexanediamine (TOYOCAT-MR, manufactured by TOSOH CORPORATION)
- 9) N,N-dimethylcyclohexylamine (manufactured by Aldrich Co.)
- 10) Crude MDI (MR-200), manufactured by Nippon Polyurethane K.K., INDEX=(mols of NCO groups/mols of OH groups) $\times 100$

Table 6
Comparative Examples

	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29
Reactivity (sec)															
Cream time	31	25	27	29	24	25	31	25	25	30	25	26	31	25	26
Gel time	90	89	90	90	91	90	90	90	91	90	90	90	90	90	90
Tack free time	131	123	125	129	122	126	130	124	125	128	126	125	129	125	126
Rise time	146	139	142	143	137	140	145	138	139	143	136	139	143	139	140
Physical properties of foam															
Flowability (cm)	81	84	83	80	83	82	81	83	83	80	84	83	81	84	84
Core density (kg/m ³)	29.2	28.2	28.4	29.3	28.3	28.0	29.0	28.2	28.4	29.3	28.4	28.3	29.3	28.0	28.1
Adhesive strength (kgf/cm ²)	1.62	1.76	1.71	1.70	1.86	1.79	1.67	1.82	1.75	1.65	1.83	1.72	1.62	1.78	1.70
Dimensional stability (%)	-1.0	-1.2	-1.3	-0.8	-1.2	-1.1	-0.8	-1.0	-1.2	-0.9	-1.3	-1.5	-0.9	-1.3	-1.2

As is evident from Tables 2, 4 and 6, it is possible to produce foams excellent in flowability, adhesive strength and dimensional stability by using amine compounds of the present invention as catalysts.

5 Namely, Examples 1 to 12 are examples wherein rigid polyurethane foams were produced by using catalysts of the present invention and 1,1,1,3,3-pentafluoropropane (HFC-245fa) as a blowing agent. In each Example, a rigid urethane foam excellent in flowability, adhesive strength
10 and dimensional stability, was obtained. Further,
Examples 13 to 24 are Examples wherein rigid polyurethane foams were produced by using the catalysts of the present invention and 1,1,1,3,3-pentafluorobutane (HFC-365mfc) as a blowing agent. In each Example, a rigid urethane foam
15 excellent in flowability, adhesive strength and dimensional stability, was obtained.

Whereas, Comparative Examples 1 to 4 and Comparative Examples 8 to 11 are examples wherein rigid polyurethane foams were produced by using only the aliphatic amine compounds of the above formula (1) as catalysts, whereby large amounts of the catalysts were required, and the foams were inferior in flowability and dimensional stability.

Further, Comparative Examples 5 to 7 and Comparative Examples 12 to 14 are examples wherein rigid polyurethane foams were produced by using triethylenediamine, N,N,N',N'-tetramethyl-1,6-hexanediamine or N,N-

dimethylcyclohexylamine solely as the catalyst without using the aliphatic amine compound of the above formula (1), whereby the obtained foams were inferior in flowability, adhesive strength and dimensional stability.

5 Further, Comparative Examples 15 to 21 are examples in which 1,1-dichloro-1-fluoroethane (HCFC-141b) was used as a blowing agent, whereby even if the catalyst of the present invention was used, no remarkable effect was observed in the flowability, adhesive strength and 10 dimensional stability of the foams.

EXAMPLES 25 to 48

A polyol, a blowing agent and a foam stabilizer were mixed in a blend ratio as identified in Table 7 to prepare Premix A. 47.1 g of Premix A was taken into a 15 300 ml polyethylene cup, and a catalyst as identified in Table 7 was added in an amount such that the reactivity as represented by the following gel time would be 90 seconds, whereupon the temperature was adjusted to 20°C. A polyisocyanate liquid (MR-200) having the temperature 20 adjusted to 10°C in a separate container, was put into the cup of Premix A in such an amount that the isocyanate index [isocyanate groups/OH groups (molar ratio) × 100)] would be 110, and immediately stirred by a stirrer at 6,500 rpm for 5 seconds. The mixed and stirred liquid 25 was transferred to a 2 liter polyethylene cup having the temperature adjusted to 40°C, and the reactivity during foaming was measured. Then, the starting material scale

was increased, and in the same manner, the mixed liquid was put into a mold having the temperature adjusted to 40°C, whereupon foam molding was carried out. Upon expiration of 10 minutes from the introduction of the 5 mixed liquid, the foam was removed from the mold. From the molded foam, the flowability, core density, adhesive strength and dimensional stability of the foam were evaluated. The results are shown in Table 8.

Table 7

Table 7 (continued)

	Blend (parts by weight)	Examples										
		38	39	40	41	42	43	44	45	46	47	48
Polyol A ¹⁾	80	80	80	80	80	80	80	80	80	80	80	80
Polyol B ²⁾	20	20	20	20	20	20	20	20	20	20	20	20
Cyclopentane	10	10	10	10	10	10	10	10	10	10	10	10
Isopentane	4	4	4	4	4	4	4	4	4	4	4	4
Water	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Foam stabilizer ³⁾	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Catalyst A ⁴⁾	0.23	0.33										
Catalyst B ⁵⁾		0.40	0.28	0.70								
Catalyst C ⁶⁾												
Catalyst D ⁷⁾												
Catalyst E ⁸⁾												
Catalyst F ⁹⁾	0.45											
Catalyst G ¹⁰⁾		0.65										
Isocyanate INDEX ¹¹⁾	110	110	110	110	110	110	110	110	110	110	110	110

1) Waste PET type polyester polyol (OH value=241 mgKOH/g) manufactured by Oxid Co.

2) Mannich type polyether polyol (OH value=469 mgKOH/g) manufactured by Daiichi Kogyo Seiyaku Co., Ltd.

3) Silicone type surfactant (tradename: L-5420), manufactured by Nippon Unicar Co., Ltd.

4) N,N-dimethylbutylamine (manufactured by Aldrich Co.)

5) N,N-dimethylhexylamine (manufactured by Aldrich Co.)

6) N,N-dimethyloctylamine (manufactured by Tokyo Kasei K.K.)

7) N,N-dimethyldecylamine (manufactured by Tokyo Kasei K.K.)

8) Dipropylene glycol solution containing 33 wt% of triethylenediamine (TEDA-L33, manufactured by TOSOH CORPORATION)

9) N,N,N',N'-tetramethyl-1,6-hexanediamine (TOYOCAT-MR, manufactured by TOSOH CORPORATION)

10) N,N-dimethylcyclohexylamine (manufactured by Aldrich Co.)

11) Crude MDI (MR-200), manufactured by Nippon Polyurethane K.K., INDEX=(mols of NCO groups/mols of OH groups) $\times 100$

Table 8

	Examples											
	25	26	27	28	29	30	31	32	33	34	35	36
Reactivity (sec)												
Cream time	30	25	26	29	25	26	31	25	25	30	26	26
Gel time	90	90	91	90	89	90	91	90	90	91	90	90
Tack free time	150	143	143	148	143	143	150	143	144	148	145	144
Rise time	161	147	153	155	148	151	158	148	150	157	148	153
Physical properties of foam												
Flowability (cm)	78	79	80	76	78	78	76	80	79	76	80	79
Core density (kg/m ³)	31.6	30.4	30.6	31.6	30.6	30.7	31.3	30.3	30.6	31.7	30.7	30.4
Adhesive strength (kgf/cm ²)	1.64	1.81	1.68	1.85	2.03	1.88	1.69	1.86	1.72	1.6	1.79	1.71
Dimensional stability (%)	-0.9	-1.2	-1.1	-0.6	-1.1	-1.0	-0.7	-1.0	-1.1	-0.8	-1.2	-1.4

Table 8 (continued)

		Examples											
		37	38	39	40	41	42	43	44	45	46	47	48
Reactivity (sec)													
Cream time	29	24	25	28	24	25	30	24	24	29	25	25	25
Gel time	89	90	90	91	90	90	90	89	89	91	90	90	90
Tack free time	151	140	145	148	142	143	149	143	143	149	143	143	144
Rise time	162	145	150	159	148	151	155	145	150	158	146	148	
Physical properties of foam													
Flowability (cm)	79	80	81	77	79	79	77	81	80	77	81	81	80
Core density (kg/m ³)	30.7	29.5	29.7	30.7	29.7	29.8	30.4	29.4	29.7	30.8	29.8	29.8	29.5
Adhesive strength (kgf/cm ²)	1.59	1.76	1.63	1.80	1.97	1.82	1.64	1.80	1.67	1.55	1.74	1.74	1.66
Dimensional stability (%)	-0.8	-1.1	-1.0	-0.5	-1.0	-0.9	-0.6	-0.9	-1.0	-0.7	-1.1	-1.1	-1.3

COMPARATIVE EXAMPLES 30 to 43

A polyol, a blowing agent and a foam stabilizer were mixed in a blend ratio as identified in Table 9 to prepare Premix A. 47.1 g of Premix A was taken into a 5 300 ml polyethylene cup, and a catalyst as identified in Table 9 was added in an amount such that the reactivity as represented by the following gel time would be 90 seconds, whereupon the temperature was adjusted to 20°C. A polyisocyanate liquid (MR-200) having the temperature 10 adjusted to 20°C in a separate container, was put into the cup of Premix A in such an amount that the isocyanate index [isocyanate groups/OH groups (molar ratio) × 100] would be 110, and immediately stirred by a stirrer at 6,500 rpm for 5 seconds. The mixed and stirred liquid 15 was transferred to a 2 liter polyethylene cup having the temperature adjusted to 40°C, and the reactivity during foaming was measured. Then, the starting material scale was increased, and in the same manner, the mixed liquid was put into a mold having the temperature adjusted to 20 40°C, whereupon foam molding was carried out. Upon expiration of 10 minutes from the introduction of the mixed liquid, the foam was removed from the mold. From the molded foam, the flowability, core density, adhesive strength and dimensional stability of the foam were 25 evaluated. The results are shown in Table 10.

Table 9

	Blend (parts by weight)	Comparative Examples											
		30	31	32	33	34	35	36	37	38	39	40	41
Polyol A ¹⁾	80	80	80	80	80	80	80	80	80	80	80	80	80
Polyol B ²⁾	20	20	20	20	20	20	20	20	20	20	20	20	20
Cyclopentane	16	16	16	16	16	16	16	16	10	10	10	10	10
Isopentane									4	4	4	4	4
Water	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Foam stabilizer ³⁾	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Catalyst A ⁴⁾	1.60							1.45		1.65			
Catalyst B ⁵⁾		1.85								1.90			
Catalyst C ⁶⁾			2.10				2.55				2.30		
Catalyst D ⁷⁾							1.55		1.10				
Catalyst E ⁸⁾									1.45				
Catalyst F ⁹⁾													
Catalyst G ¹⁰⁾													
Isocyanate INDEX ¹¹⁾	110	110	110	110	110	110	110	110	110	110	110	110	110

1) Waste PET type polyester polyol (OH value=241 mgKOH/g) manufactured by Oxid Co.

2) Mannich type polyether polyol (OH value=469 mgKOH/g) manufactured by Daiichi Kogyo Seiyaku Co., Ltd.

3) Silicone type surfactant (tradename: L-5420), manufactured by Nippon Unicar Co., Ltd.

4) N,N-dimethylbutylamine (manufactured by Aldrich Co.)

5) N,N-dimethylhexylamine (manufactured by Aldrich Co.)

6) N,N-dimethyloctylamine (manufactured by Tokyo Kasei K.K.)

7) N,N-dimethyldecylamine (manufactured by Tokyo Kasei K.K.)

8) Dipropylene glycol solution containing 33 wt% of triethylenediamine (TEDA-L33, manufactured by TOSOH CORPORATION)

9) N,N,N',N'-tetramethyl-1,6-hexanediamine (TOYOCAT-MR, manufactured by TOSOH CORPORATION)

10) N,N-dimethylcyclohexylamine (manufactured by Aldrich Co.)

11) Crude MDI (MR-200), manufactured by Nippon Polyurethane K.K., INDEX=(mols of NCO groups/mols of OH groups) × 100

Table 10

		Comparative Examples													
		30	31	32	33	34	35	36	37	38	39	40	41	42	43
Reactivity (sec)		25	24	26	25	30	25	26	24	24	25	24	29	24	25
Cream time		90	91	90	90	89	90	90	90	90	90	90	89	90	90
Gel time		145	143	148	142	150	140	143	143	143	146	140	151	143	141
Tack free time		156	157	159	153	162	155	160	155	156	157	155	160	158	156
Rise time															
Physical properties of foam		73	74	72	73	70	73	73	74	75	73	74	70	74	74
Flowability (cm)		32.2	32.1	32.2	32.0	33.5	32.1	32.2	31.9	31.8	31.9	31.6	33.2	31.8	31.9
Core density (kg/m ³)		1.79	2.03	1.95	1.83	0.90	1.02	0.96	1.74	1.95	1.88	1.78	0.88	1.00	0.95
Adhesive strength (kgf/cm ²)		-7.7	-8.1	-7.5	-6.8	-3.1	-4.6	-5.0	-6.8	-7.2	-6.8	-6.0	-2.8	-4.3	-4.3
Dimensional stability (%)															

COMPARATIVE EXAMPLES 44 to 58

A polyol, a blowing agent and a foam stabilizer were mixed in a blend ratio as identified in Table 11 to prepare Premix A. 47.1 g of Premix A was taken into a 5 300 ml polyethylene cup, and a catalyst as identified in Table 11 was added in an amount such that the reactivity as represented by the following gel time would be 90 seconds, whereupon the temperature was adjusted to 20°C. A polyisocyanate liquid (MR-200) having the temperature 10 adjusted to 20°C in a separate container, was put into the cup of Premix A in such an amount that the isocyanate index [isocyanate groups/OH groups (molar ratio) $\times 100$] would be 110, and immediately stirred by a stirrer at 6,500 rpm for 5 seconds. The mixed and stirred liquid 15 was transferred to a 2 liter polyethylene cup having the temperature adjusted to 40°C, and the reactivity during foaming was measured. Then, the starting material scale was increased, and in the same manner, the mixed liquid was put into a mold having the temperature adjusted to 20 40°C, whereupon foam molding was carried out. Upon expiration of 10 minutes from the introduction of the mixed liquid, the foam was removed from the mold. From the molded foam, the flowability, core density, adhesive strength and dimensional stability of the foam were 25 evaluated. The results are shown in Table 12.

Table 11

Blend (parts by weight)	Amine catalyst	Comparative Example														
		44	45	46	47	48	49	50	51	52	53	54	55	56	57	58
Polyol A ¹⁾		80	80	80	80	80	80	80	80	80	80	80	80	80	80	80
A Polyol B ²⁾		20	20	20	20	20	20	20	20	20	20	20	20	20	20	20
HCFC-141b		25	25	25	25	25	25	25	25	25	25	25	25	25	25	25
Water		1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Foam stabilizer ³⁾		1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Catalyst A ⁴⁾	0.35	0.25	0.35													
Catalyst B ⁵⁾				0.45	0.33	0.40										
Catalyst C ⁶⁾							0.50	0.30	0.45							
Catalyst D ⁷⁾								1.0		0.60	0.60	0.40	0.55			
Catalyst E ⁸⁾		0.70		0.50		0.90				0.80	0.90	1.20				
Catalyst F ⁹⁾					0.70							0.80		1.10		
Catalyst G ¹⁰⁾															1.10	1.45
Isocyanate INDEX ¹¹⁾	110	110	110	110	110	110	110	110	110	110	110	110	110	110	110	110

1) Waste PET type polyester polyol (OH value=241 mgKOH/g) manufactured by Oxide Co.
 2) Mannich type polyether polyol (OH value=469 mgKOH/g) manufactured by Daiichi Kogyo Seiyaku Co., Ltd.
 3) Silicone type surfactant (tradename: L-5420), manufactured by Nippon Unicar Co., Ltd.
 4) N,N-dimethylbutylamine (manufactured by Aldrich Co.)
 5) N,N-dimethylhexylamine (manufactured by Aldrich Co.)
 6) N,N-dimethyloctylamine (manufactured by Tokyo Kasei K.K.)
 7) N,N-dimethyldecylamine (manufactured by Tokyo Kasei K.K.)
 8) Dipropylene glycol solution containing 33 wt% of triethylenediamine (TEDA-L33, manufactured by TOSOH CORPORATION)
 9) N,N,N',N'-tetramethyl-1,6-hexanediamine (TOYOCAT-MR, manufactured by TOSOH CORPORATION)
 10) N,N-dimethylcyclohexylamine (manufactured by Aldrich Co.)
 11) Crude MDI (MR-200), manufactured by Nippon Polyurethane K.K., INDEX=(mols of NCO groups/mols of OH groups) × 100

Table 12

	Comparative Examples														
	44	45	46	47	48	49	50	51	52	53	54	55	56	57	58
Reactivity (sec)															
Cream time	30	24	25	28	24	30	24	25	25	29	24	25	30	24	25
Gel time	90	90	91	90	89	90	91	90	90	89	90	90	91	91	90
Tack free time	152	143	145	150	143	144	150	144	145	148	143	145	150	145	146
Rise time	166	139	142	143	137	140	145	138	139	143	136	139	143	139	140
Physical properties of foam															
Flowability (cm)	78	81	80	77	80	79	78	80	80	77	81	80	78	81	81
Core density (kg/m ³)	30.4	29.4	29.6	30.5	29.5	29.2	30.2	29.4	29.6	30.5	29.6	29.5	30.5	29.2	29.3
Adhesive strength (kgf/cm ²)	1.64	1.78	1.73	1.72	1.88	1.81	1.69	1.84	1.77	1.67	1.85	1.74	1.64	1.80	1.72
Dimensional stability (%)	-1.1	-1.4	-1.5	-0.9	-1.3	-0.1	-0.9	-1.1	-1.3	-0.9	-1.4	-1.5	-1.0	-1.4	-1.3

As is evident from Tables 8, 10 and 12, it is possible to produce foams excellent in flowability, adhesive strength and dimensional stability by using amine compounds of the present invention as catalysts.

5 Namely, Examples 25 to 48 are examples wherein rigid polyurethane foams were produced by using low boiling point hydrocarbons having boiling points of from -30 to 90°C as blowing agents. In each Example, a rigid urethane foam excellent in flowability, adhesive strength 10 and dimensional stability, was obtained.

Whereas, Comparative Examples 30 to 33 and Comparative Examples 37 to 40 are examples wherein rigid polyurethane foams were produced by using only the aliphatic amine compounds of the above formula (1) as 15 catalysts, whereby large amounts of the catalysts were required, and the foams were inferior in flowability and dimensional stability.

Further, Comparative Examples 34 to 36 and Comparative Examples 41 to 43 are examples wherein rigid 20 polyurethane foams were produced by using triethylenediamine, N,N,N',N'-tetramethyl-1,6-hexanediamine or N,N-dimethylcyclohexylamine solely as the catalyst without using the aliphatic amine compound of the above formula (1), whereby the foams were inferior 25 in flowability, adhesive strength and dimensional stability.

Further, Comparative Examples 44 to 58 are examples

in which 1,1-dichloro-1-fluoroethane (HCFC-141b) was used as a blowing agent, whereby even if the catalyst of the present invention was used, no distinct effect to the flowability, adhesive strength and dimensional stability, 5 was observed.

EXAMPLES 49 to 52 and COMPARATIVE EXAMPLES 59 to 61

A polyol, a blowing agent and a foam stabilizer were mixed in a blend ratio as identified in Table 13 to prepare Premix A. 47.1 g of Premix A was taken into a 10 300 ml polyethylene cup, and a catalyst as identified in Table 13 was added in an amount such that the reactivity as represented by the following gel time would be 90 seconds, whereupon the temperature was adjusted to 10°C. A polyisocyanate liquid (MR-200) having the temperature 15 adjusted to 10°C in a separate container, was put into the cup of Premix A in such an amount that the isocyanate index [isocyanate groups/OH groups (molar ratio) × 100] would be 110, and immediately stirred by a stirrer at 6,500 rpm for 5 seconds. The mixed and stirred liquid 20 was transferred to a 2 liter polyethylene cup having the temperature adjusted to 40°C, and the reactivity during foaming was measured. Then, the starting material scale was increased, and in the same manner, the mixed liquid 25 was put into a mold having the temperature adjusted to 40°C, whereupon foam molding was carried out. Upon expiration of 10 minutes from the introduction of the mixed liquid, the foam was removed from the mold. From

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Appeal Brief

RELATED PROCEEDINGS APPENDIX

None.